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Disinfection for Potable Reuse

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16. ABSTRACT

Potable reuse requires a multiple barrier approach to control pathogens from a grossly contaminated source. The primary multiple pathogen barriers used at the Denver Potable Water Reuse Demonstration Plant are excess lime treatment, ozonation, reverse osmosis, and chlorine dioxide. Undisinfected secondary wastewater effluent is subjected to high-pH lime treatment which inactivates viruses and bacteria, reduces TOC, removes suspended solids, and precipitates heavy metals and phosphorus. Ozone is an effective virucide and bactericide and chemically oxidizes refractory organics. Reverse osmosis is a physical barrier to pathogens and removes ions, certain organics, and nitrogen forms. Chlorine dioxide provides a persistent residual and minimizes the potential for chloro-organic formation. Operational experience is described and disinfection performance evaluated.

17. KEY WORDS AND DOCUMENT ANALYSIS

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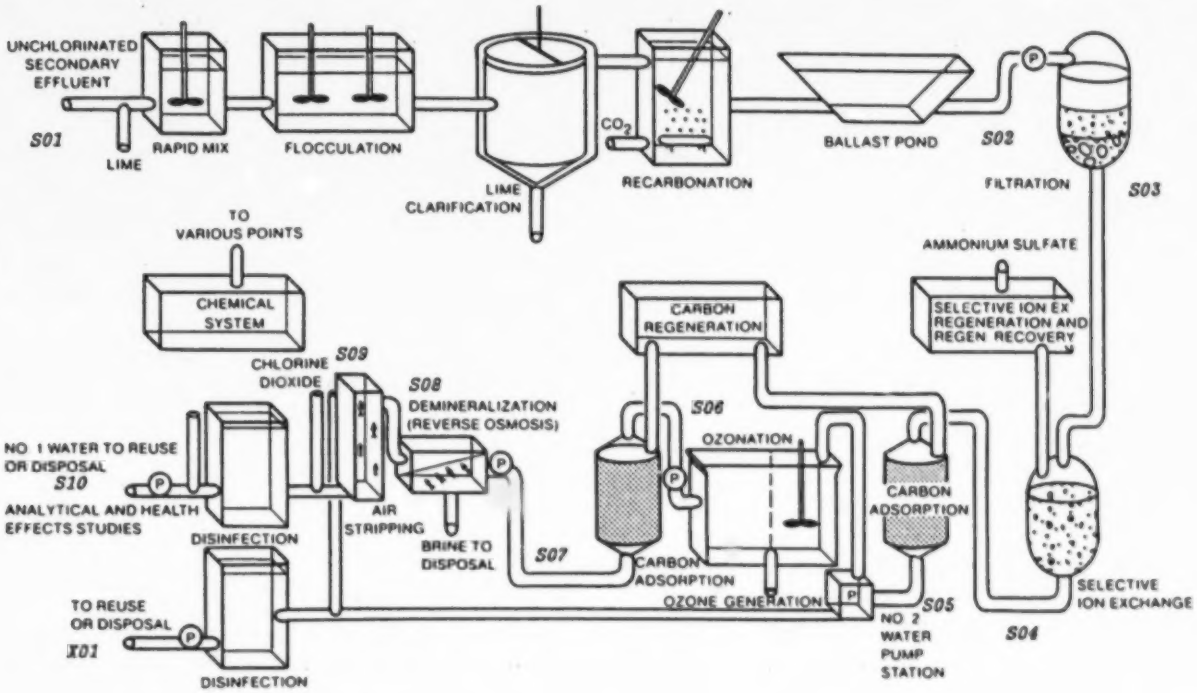

Figure 1–Water reuse treatment process.

METHODS

Bacteria. Total coliforms (TC), fecal coliforms (FC), fecal streptococci (FS) and the heterotrophic plate count (HPC) were analyzed by membrane filtration methods according to the 16th edition of "Standard Methods," methods 909A, 909C, 910B, and 907C, respectively.[11] The membrane heterotrophic plate count (m-HPC) has supplanted the traditional standard plate count (SPC) earlier because of better recovery, increased sample volume, and higher precision.[12]

Coliphage. Bacteriophages of *E. coli* B and *E. coli* C-3000 (coliphages) were analyzed according to Grabow *et al.*[10] The method has now been modified to improve recoveries so that 5.0 *mM* Ca^{+2} and 1.25 *mM* Mg^{+2} are added,[13] and samples are filtered through two 1 MDS Virosorb electropositive membrane filters (AMF Cuno, Meridian, CN) that are eluted with a 4.0% beef extract plus 0.5 *M* NaCl at pH 9.00.[14] This modified technique has allowed better coliphage recoveries than with method 919C of "Standard Methods."[11] Dee[15] recovered only 2.1% of coliphage *E. coli* B, and 13.2% of coliphage *E. coli* C by method 919C compared to this modified technique (geometric mean values).

Chemistry. Ozone concentration in air was measured continuously by ultraviolet absorption. Ozone residual in water was continuously measured by polarographic electrode; laboratory corroboration was measured by a N,N-diethyl-p-phenylene diamene (DPD) spectrophotometric method. Chlorine dioxide residual was continuously measured by electrode, and was corroborated by a modified DPD method ("Standard Methods," method 410C);[11] chlorine dioxide speciation was measured amperometrically according to Aieta, *et al.*[16] All process-stream monitoring sensors were corroborated with laboratory test results at 4-hour intervals.

Data. Data were accumulated, stored, and processed by the plant computer system. The capabilities of this system are discussed elsewhere.[17] The statistical methods used in data analysis (for example, lognormal statistics) were developed to work in situations where some data is effectively censored from view when below detectable levels.[18] The integrity of the data is ensured by adherence to quality assurance (QA) procedures contained in manuals developed for the project.

RESULTS AND DISCUSSION

Results are for the 6-month final shakedown of the plant after reverse osmosis became operational (March 1 to September 1, 1985). The data represent full-scale operation under the conditions of variable influent quality, pH setpoints, ozone dosages, reverse osmosis recoveries, and chlorine dioxide dosages. Results across the plant are summarized in Table 1 and are graphically represented in Figures 2 through 6. It is important to note that the data represent a lengthy and highly variable start-up period. A minimum of 5 years of intensive study[4] will confirm or negate the preliminary conclusions drawn.

Lime treatment. The reduction of total coliforms, heterotrophic plate count organisms, and coliphages is presented in Table 1. Total coliforms were reduced by a factor of 3.37 logs, or 99.96% removal at pH 10.6 to 11.6 (Figure 2). The hetero-

Table 1—Microorganism counts (geometric means), March 1–September 1, 1985.

Microorganism	S01	S02	S05	S06	S07	S08	S09	S10	S12
Total coliform/100 mL	4.32×10^5	184.78	78.81	2.58	8.16	0.62	0.92	2.52×10^{-6}	0.001
Log count/100 mL	5.64	2.27	1.90	0.41	0.91	−0.21	−0.03	−5.6	−3.0
m-HPC/mL	1.44×10^5	1.37×10^5	5.18×10^4	2.70×10^4	4.45×10^4	135.72	33.59	0.25	0.46
Log count/mL	5.16	5.14	4.71	4.43	4.65	2.13	1.53	−0.60	−0.34
E. coli B PFU/100 mL	1.69×10^4	4.51	3.45	ND	2.00×10^{-5}	ND	ND	ND	ND
Log count/100 mL	4.23	0.65	0.54	ND	−4.70	ND	ND	ND	ND
E. Coli C-3000 PFU/100 mL	3.81×10^4	7.58	4.70	1.57×10^{-6}	2.20×10^{-6}	ND	ND	ND	ND
Log count/100 mL	4.58	0.88	0.67	−5.80	−5.66	ND	ND	ND	ND
Fecal coliform/100 mL	8.45×10^3	—	—	—	—	—	—	ND	ND
Log count/100 mL	3.93	—	—	—	—	—	—	ND	ND
Fecal streptococci/100 mL	2.79×10^3	—	—	—	—	—	—	ND	ND
Log count/100 mL	3.44	—	—	—	—	—	—	ND	ND

Note—Sample locations are illustrated in Fig. 1. S12 is the Denver drinking water comparison sample. ND = Not detected.

trophic plate count populations were little affected following lime treatment, however, and were only slightly reduced in numbers. This is in contrast to the South African reuse experience,[10] which showed a total plate count reduction of 99.44% at pH 11.1 (range of 10.9 to 11.3). Because the lime treatment samples are from a point (S02, Figure 1) downstream of recarbonation (approximately pH 7.7) and the ballast pond (hydraulic residence time of approximately 5 hours), it is not known whether these data reflect microorganism resistance to the high pH environment, a difference between total and heterotrophic plate count methodologies, or a regrowth of microorganisms.

Coliphages were analyzed as a model for enteric virus removals. Coliphages were reduced by a factor of 3.58 logs, or 99.97% removal for the *E. coli* B Host, and by a factor of 3.70 logs, or 99.98% removal for the *E. coli* C-3000 Host. These results are encouraging for projecting removals at enteric viruses, which have been reported to be more sensitive to lime treatment than coliphages.[10] Thus, for lime treatment at least, coliphages are apparently conservative models for enteric virus removal.

Ozone. Ozonation has two objectives: it is the first disinfectant applied to the process flow, and it is an oxidant that may modify organic compounds and improve their adsorbability by granular activated carbon (GAC) and their biodegradability by the biological activated carbon (BAC) bed that follows.[19-21] These two objectives can be at cross-purposes, however. Residual ozone applied to second-stage carbon has the potential to degrade the carbon matrix;[22] therefore, the ozone process is operated to maintain a minimum detectable residual after approximately 60 minutes of contact. Ozone data is reported for a countercurrent application.

Total coliform removal was 1.49 logs, or a 96.73% reduction. Applied dosage over the specified time period varied from 2.12 mg/L to 3.94 mg/L. Transfer efficiencies ranged from 80.8% to 87.7%, and absorbed dosages varied from 1.76 mg/L to 3.26 mg/L. Relating log removal of total coliform (Log N_o/N) to log absorbed dose, as described by Venosa[23] and Venosa and Opatken[24], yields a correlation coefficient, $r = 0.77$ as shown in Figure 3. N_o represents ozone influent total coliform count, and N represents ozone effluent total coliform count. The least-square regression line is described by:

$$\text{Log } (N_o/N) = 4.73 \text{ Log (absorbed dose)} - 1.02 \qquad (1)$$

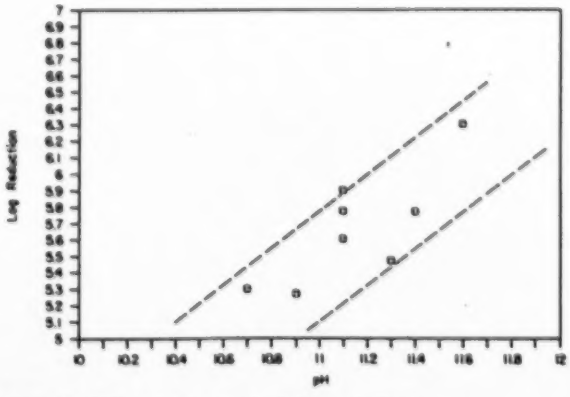

Figure 2—Effect of pH on total coliform reduction.

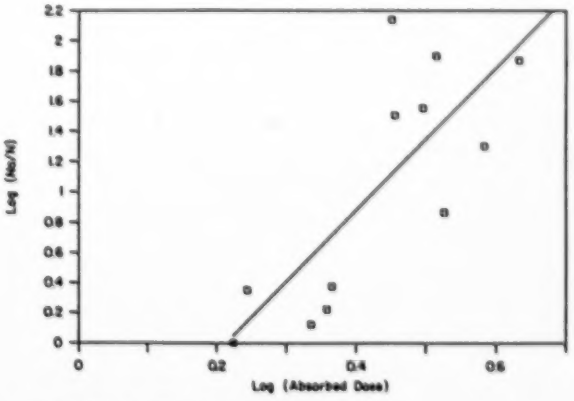

Figure 3—Total coliform reduction by ozone.

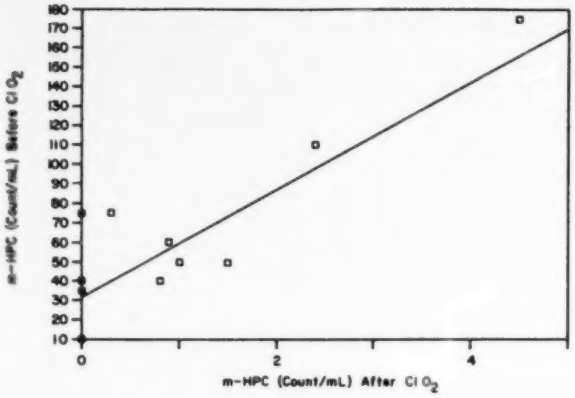

Figure 4—Effect of disinfection of m-HPC.

The only data plotted are those for nonzero values of N; that is, if there are no detectable total coliforms in the ozone effluent (S06), the point was not used for correlation purposes.

Heterotrophic plate count organisms were less sensitive to ozone than coliforms; log removal was 0.28, or a 47.88% reduction. Coliphages, already reduced to low levels by lime treatment, were essentially undetectable following ozonation (Table 1).

Reverse osmosis. Reverse osmosis (RO) is the last unit process before final disinfection. The spiral-wound polyamide membranes are a physical barrier to the passage of pathogens. The process removes toxic metals, some nitrogen forms, and certain organics.[4,25]

Total coliforms were reduced by 1.12 logs, or 92.40% removal. Plate count was reduced by 2.52 logs, or 99.70% removal. Coliphages were never detected following reverse osmosis. Ideally, pathogens should be totally absent from RO product water. An explanation for the occasional recovery of total coliforms from RO product water (S08), in 22 out of 57 samples, was discovered during inspection of the modules. Brine seals were cocked so as to allow some short-circuiting of the flow stream. Also, some product tubes connecting the modules were fractured, which provided a potential for microorganism short-circuiting and colonization. This apparently was caused by the compression and decompression of the modules during system start-up. The seals and fractured modules were replaced and spacers were added between the modules to minimize movement during pressurization/depressurization. This retrofit, when applied to all of the modules, should further reduce microorganism counts in RO product water.

Chlorine dioxide. Chlorine dioxide (ClO_2) was selected as the final disinfectant to provide a persistent residual while minimizing the formation of chlorinated organic compounds. Chlorine dioxide is generated by reacting an acidified chlorine solution (pH 2 to 3) with sodium chlorite ($NaClO_2$) to produce a chlorine dioxide solution that is then pumped to the final product water, W1 (S10), and the intermediate process water, W2 (X01), which is used for irrigation and for backwash of filters, ion exchange columns, and first-stage carbon adsorption beds. The generator can produce high yields consistently more than 95% (based on $NaClO_2$ feed) with minimum byproducts of chlorite (ClO_2^-) and chlorate (ClO_3^-), which are significant to human health.[26] The generator has been operated at solution strengths less than 500 mg/L to meet the low dosage requirements for W1 and W2. This has required the addition of excess chlorine to drive the reaction to completion.[27] Excess chlorine has not been required at higher solution strengths. A retrofit project is presently underway that will provide storage of the chlorine dioxide solution to obviate the excess chlorine requirement at existing demand levels. Also, new generators will be tested in the near future.

Total coliform mean values, which were significantly reduced after reverse osmosis, were further reduced by more than 5 logs; however, it must be noted that this level of reduction is a product of statistical analysis. Mean plate counts were substantially reduced by chlorine dioxide—a 2.13 log reduction, or 99.26% removal. Figure 4 relates the effect of chlorine dioxide on m-HPC organisms. The least-square regression expression (correlation coefficient $r = 0.92$) is

$$\text{m-HPC}_{(\text{after})} = 0.0356\ \text{m-HPC}_{(\text{before})} - 1.10 \qquad (2)$$

The m-HPC counts for both axes are significant when compared to the recommended drinking water level of 500/mL. m-HPC counts before chlorine dioxide disinfection ranged to a high of 180/mL while the maximum count after chlorine dioxide application was 5/mL. ClO_2 dosages for the data in Table 1 ranged from 0.10 to 1.30 mg/L, with an average of 0.49 mg/L; residuals ranged from 0.05 mg/L to 0.68 mg/L, with an average of 0.24 mg/L.

SUMMARY

Total coliforms. Total coliform removal is depicted in Figure 5. The six month geometric mean value was below the National Primary Drinking Water Maximum Contaminant Level (MCL) of 1/100 mL (log 0) following reverse osmosis. Chlorine dioxide disinfection further reduces this level to essentially zero, which provides a significant safety factor. The influent population of 5.64 log counts/100 mL was reduced by a factor of 5.61 logs before final disinfection. Fecal coliforms and fecal streptococci present in substantial numbers in the influent (3.93 and 3.44 log counts/100 mL, respectively) were never detected in the plant product water.

m-HPC. Heterotrophic plate count organisms were somewhat more resistant than the coliforms. Their removal across the plant is shown in Figure 6. Regrowth is demonstrated by the increase

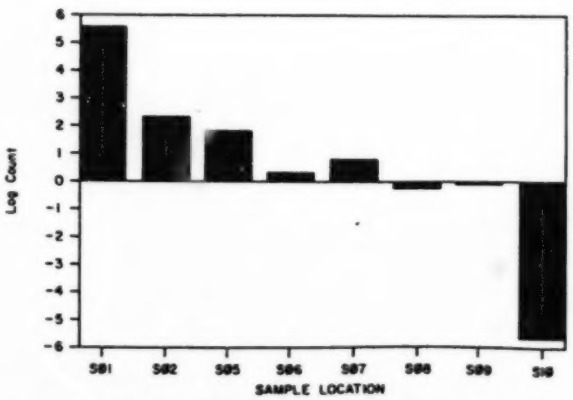

Figure 5—Total coliform (mean log count/100 mL).

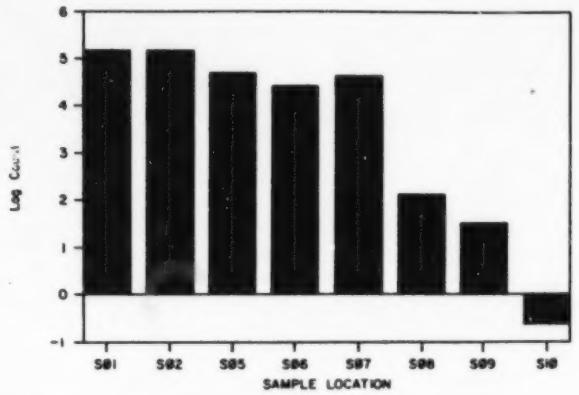

Figure 6—m-HPC removal (mean log count/mL).

in organisms following ozonation, and may be responsible for the apparent ineffectiveness of lime treatment. The six month geometric mean value was below the National Secondary Drinking Water Recommended Maximum Contaminant Level (RMCL) of 500/mL (log 2.7) following reverse osmosis. Chlorine dioxide further reduced these counts by more than 2 logs, again providing a significant safety factor.

Coliphages. Coliphage *E. coli* B and *E. coli* C-3000 removal across the plant is presented in Table 1. Lime treatment is very effective in reducing the coliphage population by factors of more than 3.5 logs. Coliphages were essentially undetectable following ozonation. While coliphages are only a model for enteric virus removal, this performance is very encouraging. Enteric virus analysis will be a routine part of the future plant analytical program.[27]

CONCLUSIONS

The goal of the reuse demonstration plant is to produce a water of equal or better quality than the excellent existing potable water supply for Denver.[1-4] One measure of water quality is the presence or absence of pathogenic organisms. A comparison of reuse product water (S10) with Denver drinking water (S12) mean values listed in Table 1 shows that this goal can be achieved using bacterial indicators and coliphage, even during the extreme variability of start-up conditions. A statistical test (*t*-test)[17] to determine the significance of the difference between six month mean values indicated that the values were, in fact, different at $p = 0.05$ for both TC and HPC.

The production of a water of lower bacterial count than potable water produced from first-use mountain runoff has been demonstrated by using the multiple pathogen barriers of lime treatment, ozone, reverse osmosis, and chlorine dioxide disinfection. Although meeting existing drinking water standards does not ensure potability,[7] the performance thus far indicates that a large safety factor exists for public health protection.

Future studies will include the analysis of additional pathogenic organisms such as *Giardia, Entamoeba, Salmonella, Campylobacter, Shigella,* and enteric viruses.[27] The results of these studies along with the continued monitoring for the various organisms discussed in this paper will substantiate the proposition that water reclaimed from secondary wastewater effluent can exceed the highest standards.

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